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Production and Characterization of Biodegradable Plastics based on Starch of *Artocarpus heterophyllus* Lam Seeds

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Abstract. The starch of *artocarpus heterophyllus* lam seeds was mixed with distilled water into a solution. The biodegradable plastics were made of starch, glycerol, and chitosan on the different volume fraction of glycerol at mixing temperature 80°C. Samples were prepared based on the different volume fraction of glycerol, i.e., 2, 4 and 6 (vol.%). The crystal structures of biodegradable plastic films were investigated using x-ray diffractometer and their thermal properties were analyzed employing thermogravimetric analysis. The biodegradabilities of specimens were obtained by soil burial test method using microorganisms. The maximum mechanical strength of biodegradable plastics is obtained about 4.7 MPa.

Introduction

The synthetic plastics are still widely used as raw materials in various industries due to high strength [1,2], cheap, longer life [2], and guaranteed safety, satisfaction, and aesthetic quality [3]. The widespread use of them has caused environmental pollution and consumption of the resources of non-renewable fuels increasing [4]. Society faced difficulties in waste storage for limited locations and increasing global warming due to garbage burning [5]. Many wild animals are threatened their survival because of the emergence of toxic gases [6]. To solve this problem, the researchers have developed biodegradable plastics [7]. They utilize renewable polymeric materials such as carbohydrates, starches and proteins because of some excellencies, i.e., abundant varieties of plants, lower processing cost and easy biodegradation process [8-10]. In this research, the starch of *artocarpus heterophyllus* lam seeds is selected as raw materials [2,11]. The use of biodegradable plastics films will decrease waste recycling costs and environment problems [12].

Biodegradability is an important requirement for packaging products [13], its value depends on raw material, chemical composition of constituents, and environment. Biodegradable plastic products must meet the requirements of good mechanical strength [14] in which in comparison with conventional thermoplastic, the mechanical strength of biodegradable plastic lower. Starches can be converted into thermoplastics through heat treatment mixed with plasticizer [15]. In this study, the volume fraction was optimized and effect of glycerol addition on the microstructures, mechanical strengths, thermal properties and the biodegradation duration were investigated.

Materials and Method

Materials used consist of *artocarpus heterophyllus* lam seeds starch, chitosan, acetic acid, glycerol, and distilled water. Additionally, the used chitosan powders are with specification as follows: the degree of deacetylation 87.4% and 1% volume solubility of chitosan in acetic acid by 99.4%. The chitosan solution was pepared by ionotropic gelation method.

To get rid of sap, *artocarpus heterophyllus* lam seeds are washed with water, peeled and soaked in lime water for 1 hour. They were briefly drained and crushed using a blender to get the starch. Starch was dried for 1-2 days in the sun and filtered using a sieve T.61. Starch solution is made of mixing 5ml *artocarpus heterophyllus* lam seed starch and 50ml distilled water then stirred until it forms a suspension. The chitosan powders of 5ml added acetate acid of 0.1% forms chitosan

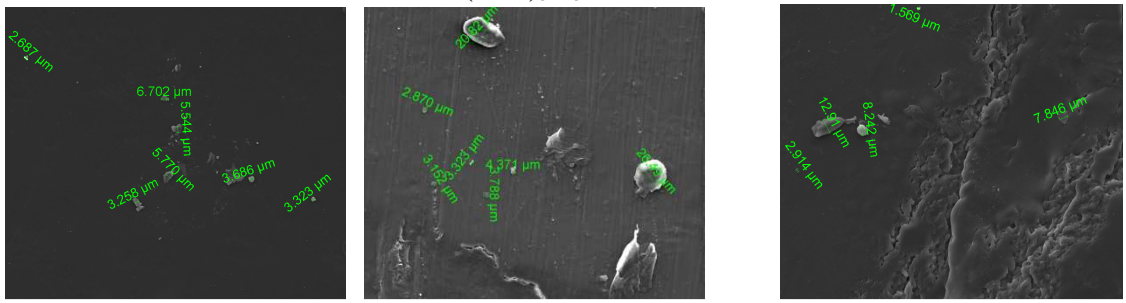
solution of 0.5%. The starch suspension of 50ml is mixed with gelatinized chitosan solution as much as 0.5ml and casted into pan in the dimension of 12.5x18.5x2 cm^3. The casted plastic materials were dried using oven (Kirin) at 45ºC for 6 hours.

The microstructures of plastics films were observed using *Scanning Electron Microscope* (SEM) (FEI, Inspect-S50) in 500x magnification of lens and operating voltage of 20kV. Specimens were prepared 1cmx1cm in size and their surfaces were coated gold (Au) using *sputter coater* (EMITECH, SC-7620). The crystal structures were characterized using diffractometer (40 kV, 100 mA) which is completed with Ni filtered Cu radiation and a curved graphite crystal monochromator at scanning speed 2º/min. Tensile test machine Testometric AX M350-10KN was used to determine mechanical properties of them, referring to testing standard ISO 1184-1983. Their thermal properties were obtained using *thermo gravimetric analyzer* (STA PT 1600) at temperature range of 30-520°C with heating rate 10°C/minute in argon atmosphere.

Results and Discussion

Microstructures play important role in determining properties of biodegradable plastics. The distribution of chitosan in the starch matrix which was plasticized using glycerol is indicated by SEM images in Figures 1(a), 1(b) and 1(c). In the specimen with lowest glycerol amount (2%Vol), the chitosan is uniformly dispersed throughout the matrix, although there was still segregation. Grain sizes of segregation range between 3.9 µm until 350 µm. The chitosan surface appears covered by a matrix because the chemical similarity of polysaccharides. In Figure 1(c), the segregation of polymers is found.

Figure 2 shows *wide angle X-ray diffraction (WAXD)* patterns for three specimens of biodegradable plastics with different glycerol quantity. The unit cell obtained is orthorombic with lattice parameters a = 9.395, b = 9.395, and c = 8.350 nm, angles of alpha = 90, beta = 90 and gamma = 120, and two dominant diffraction peaks in the fields of (012) and (110) are found clearly in angles of 2θ 23.297° and 54.8°, respectively. The sharp peak in the scattering angle of 2θ = 38.289° is also present in the diffraction pattern. Compound found in the XRD data is hydrogen carbon oxide with the chemical formula $(CH_2)_3O_3$.



(a) (b) (c)

Figure. 1. Microstures of biodegradable plastics with each glycerol volume fraction: (a) 2%, (b) 4%, dan (c) 6%.

The mechanical properties of starch-based casted products depend on the processing parameter and polymer composition. The significant decrease of mechanical strength is found in the specimen with high plasticizer amount. Figure 3 exhibits the effect of glycerol quantity on the mechanical properties of biodegradable plastics of *artocarpus heterophyllus* lam seed starch. The glycerol content affects significantly on the measured mechanical strength. The tensile strengths of specimens A(2 vol.%), B(4 vol.%), and (6 vol.%) are 5.52, 4.55 and 4.43 M.Pa, respectively, whereas the elongation at break point are 4.89, 5.90, and 5.44 mm, respectively.

The glycerol plasticization decreases hydrogen bonding. When the glycerol amount is varied between 2% and 6%, the tensile strength decreasing from 5.52 to 4.43 MPa, the elongation at break point are 4,89 to 5,90mm. This is suspected caused by interface tension between chitosan and glycerol-plasticized starch matrix, because the polysaccharide structures equal those of chitosan and starch. The chitosan can serve as junction, which increasing tensile strength, but decreasing flexibility of starch molecules. In the crack propagation theory, the pores play role as concentrators

which decreasing tensile strength of materials such as polymers and metals. The mechanical crack also proves that decreasing pore size can significantly improve tensile strength.

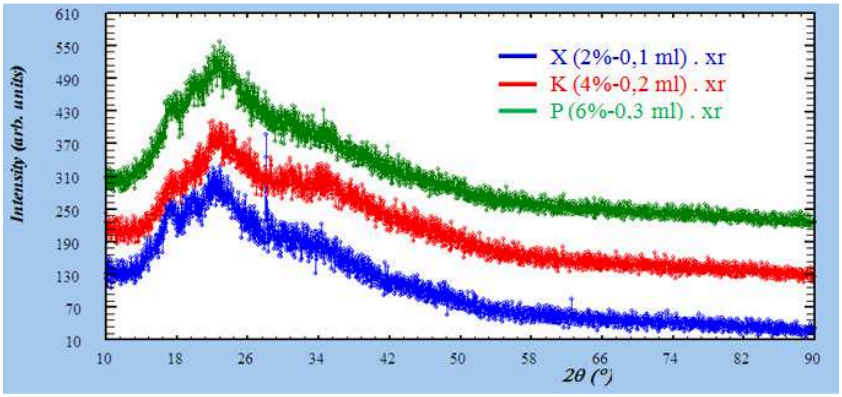


Figure. 2. WAXD patterns of biodegradable plastics with glycerol content each are: (a) 2, (b) 4, and (c) 6 vol.%.

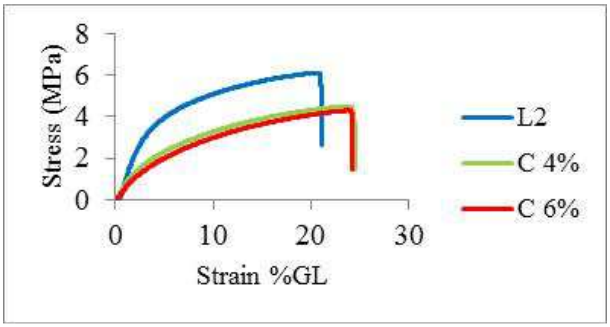


Figure. 3. Mechanical strengths of biodegradable plastics vs strains for glycerol volume fraction: (a) 2, (b) 4, and (c) 6 vol.%.

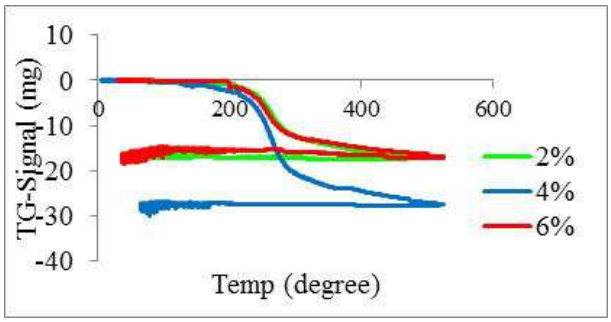


Figure. 4. TGA thermogram of three specimens with different glycerol volume fractions.

Glycerol ($C_3H_8O_3$) is a hydrophilic plasticizer with low molecular weight and can penetrate inside the pleated protein surfaces for interaction with amine acid, its plasticization effect on the mechanical strength is high and can insert inside the network of three dimension biopolymers. The glycerol effects on the tensile properties of specimens are shown in Fig. 3.

Plot of thermogravimetry data of specimens with different glycerol content is shown in Fig. 4. Heat treatment leads to mass loss under 300°C due to evaporation of liquid plasticizer as well as presence of moisture in the sheet. The second mass loss which correspond to polymer matrix degradation at temperature range of 300-500°C are between 83 and 78%. The residue mass losses for this starch are between 22 and 17%. In this study, thermogravimetric result which found for each specimen are relatively closed each other and a variation of it are found under 300°C due to evaporation of glycerol.

Biodegrability is determined to know significantly time required by specimens to completely degrade in the soil. The used method to resolve biodegradability is soil burial test, namely a direct contact method with soil by relying microorganisms in the soil. The specimens of A (2 vol.%), B(4 vol.%), and C(6 vol.%) degraded for 6, 8 and 10 days, respectively.

Conclusion

Biodegradable plastics are successfully fabricated of *artocarpus heterophyllus* lam seed starch. The polymer segregations in the microstructures are found and *carbon hydrogen oxide* could be identified as orthorombic crystal structure. The high plasticizer content has led to mechanical strength of specimen decreasing. During heat treatment process at 0-500°C has taken place mass losses twice, first at temperature range of 0-300°C and second, at 300-500°C. These specimens can be biodegraded by microorganisms in the soil for 6-10 days.

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